

# Magnetization plateau in the spin ladder with the four-spin exchange

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The magnetization process of the  $S=1/2$  antiferromagnetic spin ladder with the four-spin cyclic exchange interaction at  $T = 0$  is studied by the exact diagonalization of finite clusters and size scaling analyses. It is found that a magnetization plateau appears at half the saturation value if the ratio of the four- and two-spin exchange coupling constants  $J_4$  is larger than the critical value  $J_{4c} = 0.05 \pm 0.04$ . The phase transition with respect to  $J_4$  at  $J_{4c}$  is revealed to be the Kosterlitz-Thouless-type.

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The field-induced spin gap is one of recent interesting topics on the one-dimensional (1D) quantum spin systems. The gap can be detected as a plateau of the magnetization curve at low temperatures. The appearance of such a magnetization plateau was theoretically predicted in several systems; the anisotropic  $S = \frac{3}{2}$  antiferromagnetic chain [1,2], the  $S = 1$  bond-alternating chain [3], the  $S = \frac{1}{2}$  ferromagnetic-ferromagnetic-antiferromagnetic chain [4], the frustrated bond-alternating chain [5], the three-leg ladder [6], and the frustrated two-leg ladder [7]. In particular the two-leg ladder attracts a great interest in the context of the superconductivity in a doped system [8]. The standard  $S = \frac{1}{2}$  uniform antiferromagnetic spin ladder has the spin gap of the lowest excitation from the nonmagnetic ground state (GS), which leads to a plateau at the magnetization  $m = 0$  [9–11]. A strong coupling approach [7] indicated an additional plateau at half the saturated magnetization due to the next-nearest antiferromagnetic coupling which yields the frustration. In this paper we show another possibility of the magnetization plateau in the two-leg spin ladder, which is induced by a four-spin exchange interaction.

A four-spin exchange interaction described by a product of two-spin exchanges in a spin ladder was investigated by a field theoretical approach [12]. It indicated the possibility of a different type of massive phase from the Haldane phase [13] in the nonmagnetic GS, but the state in a strong magnetic field was not discussed. On the other hand a mean field analysis [14] suggested that the  $S = \frac{1}{2}$  triangular lattice antiferromagnet would have a magnetization plateau at half the saturated magnetization, if there exists a four-spin cyclic exchange interaction. It was verified by the exact diagonalization [15]. The recent experiments revealed that such multiple-spin exchange interactions are realized in the two-dimensional (2D) solid  $^3\text{He}$  [16,17] and the 2D Wigner solid of electrons formed in a Si inversion layer [18], as well as the bcc  $^3\text{He}$  [19]. In order to test the possibility of a similar magnetization plateau in 1D quantum spin systems, we consider the  $S = \frac{1}{2}$  uniform antiferromagnetic spin ladder with the four-spin cyclic exchange at every plaquette. The magnetization process of the system is described by

the Hamiltonian

$$\begin{aligned} \mathcal{H} &= \mathcal{H}_0 + \mathcal{H}_Z, \\ \mathcal{H}_0 &= \sum_j (\mathbf{S}_{1,j} \cdot \mathbf{S}_{1,j+1} + \mathbf{S}_{2,j} \cdot \mathbf{S}_{2,j+1} + \mathbf{S}_{1,j} \cdot \mathbf{S}_{2,j}) \\ &\quad + J_4 \sum_j (P_{4,j} + P_{4,j}^{-1}), \\ \mathcal{H}_Z &= -H \sum_j (S_{1,j}^z + S_{2,j}^z), \end{aligned} \quad (1)$$

where  $P_{4,j}$  is the cyclic permutation operator which exchanges the four spins around the  $j$ -th plaquette as  $\mathbf{S}_{1,j} \rightarrow \mathbf{S}_{1,j+1} \rightarrow \mathbf{S}_{2,j+1} \rightarrow \mathbf{S}_{2,j} \rightarrow \mathbf{S}_{1,j}$ ,  $J_4$  is the strength of the four-spin exchange and  $H$  is the applied magnetic field normalized by the two-spin exchange coupling constant. We assume  $J_4$  is positive, as it is in the solid  $^3\text{He}$ . This system subjected to the periodic boundary condition is studied by the exact diagonalization of the finite clusters and the size scaling of the low-lying energy spectra. For  $L \times 2$ -spin systems, the lowest energy of  $\mathcal{H}_0$  in the subspace where  $\sum_j (S_{1,j}^z + S_{2,j}^z) = M$  is denoted as  $E(L, M)$ . Using Lanczos' algorithm, we calculated  $E(L, M)$  ( $M = 0, 1, 2, \dots, L$ ) for even- $L$  systems up to  $L = 16$ . The macroscopic magnetization is defined as  $m = \frac{M}{L}$ .

The nonmagnetic GS of the system (1) with  $J_4 = 0$  is in a massive phase equivalent to the Haldane phase of the  $S = 1$  antiferromagnetic chain and the low-lying excitation has a finite energy gap for  $m = 0$ . On the other hand, the magnetic GS is always gapless [20,21] except for the saturation. Thus the magnetization curve has a plateau at  $m = 0$ , while no other plateau appear up to  $m = 1$ , as far as  $J_4 = 0$ . The four-spin exchange, however, is expected to induce a plateau at  $m = \frac{1}{2}$ , because the interaction stabilizes the '*uuud*' state, mentioned in Ref. [14], of the four spins around every plaquette within a mean field argument. We concentrate on the plateau at  $m = \frac{1}{2}$ , rather than the nonmagnetic GS.

The magnetic excitation gap giving  $\delta M = \pm 1$  of the  $L \times 2$ -spin systems described by the total Hamiltonian  $\mathcal{H}$  is given by

$$\Delta_{\pm} \equiv E(L, M \pm 1) - E(L, M) \mp H. \quad (2)$$

For the gapless system in the thermodynamic limit, the conformal field theory [22] (CFT) predicts the asymptotic form of the size dependence of the gap as  $\Delta_{\pm} \sim O(1/L)$  with fixed  $m = M/L$ . When  $H_+$  and  $H_-$  are defined as

$$\begin{aligned} E(L, M+1) - E(L, M) &\rightarrow H_+ \quad (L \rightarrow \infty), \\ E(L, M) - E(L, M-1) &\rightarrow H_- \quad (L \rightarrow \infty), \end{aligned} \quad (3)$$

$H_+$  and  $H_-$  has the same value and it gives the magnetic field  $H$  for the magnetization  $m$  in the thermodynamic limit. In contrast to the gapless case, if the system has a finite gap even in the infinite length limit,  $\Delta_+$  and  $\Delta_-$  are still finite for  $L \rightarrow \infty$ . It leads to the difference between  $H_+$  and  $H_-$  and a plateau appears for  $H_- < H < H_+$  at  $m = M/L$  in the magnetization curve at  $T = 0$ .

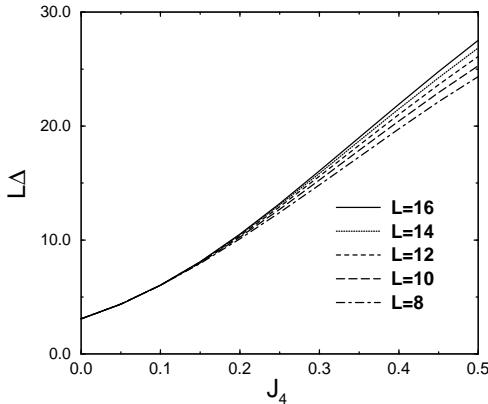


FIG. 1. Scaled gap  $L\Delta$  versus the strength of the four-spin exchange interaction  $J_4$ .

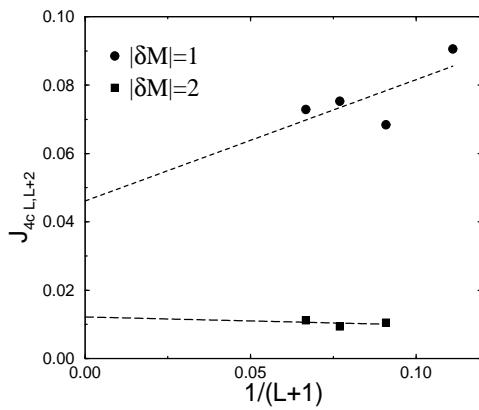


FIG. 2.  $L$ -dependent fixed point  $J_{4cL,L+2}$  of the gap for  $\delta M = \pm 1$  (circles) and  $\delta M = \pm 2$  (squares) are plotted versus  $1/(L+1)$  to determine  $J_{4c}$  in the thermodynamic limit. The estimated value is  $J_{4c} = 0.05 \pm 0.01$  for  $\delta M = \pm 1$ , which does not well agree with the result for  $\delta M = \pm 2$   $J_{4c} = 0.01 \pm 0.01$ . Thus we conclude  $J_{4c} = 0.05 \pm 0.04$ .

The sum  $\Delta \equiv \Delta_+ + \Delta_-$  is a good order parameter to investigate the plateau-nonplateau transition with the finite-size scaling [2], because  $\Delta$  corresponds to the length of the plateau in the magnetization curve in the thermodynamic limit. The scaled gap  $L\Delta$  of finite systems ( $L = 8 \sim 16$ ) at  $m = 1/2$  is plotted versus  $J_4$  in Fig. 1. For  $J_4 > 0.2$  the scaled gap obviously increases with increasing  $L$ , which means that a finite gap exists in the thermodynamic limit. For small  $J_4$  around the region  $0 < J_4 < 0.1$ , the scaled gap looks almost independent of  $L$ . It implies that the system is gapless at a finite region of the parameter  $J_4$ , which is reminiscent of the Kosterlitz-Thouless (KT) transition [23]. According to our precise analysis, the  $L\Delta$  curves for  $L$ , and  $L+2$  have an intersection in the region  $0 < J_4 < 0.1$  for each  $L$ . Thus the critical point  $J_{4c}$  can be estimated by the phenomenological renormalization group (PRG) equation [24]

$$(L+2)\Delta_{L+2}(J'_4) = L\Delta_L(J_4). \quad (4)$$

We define  $J_{4cL,L+2}$  as the  $L$ -dependent fixed point of (4) and it is extrapolated to the thermodynamic limit.  $J_{4cL,L+2}$  is plotted versus  $1/(L+1)$  as solid circles in Fig. 2. Although the convergence of  $J_{4cL,L+2}$  with increasing  $L$  is not good, the least square fitting of the form  $J_{4cL,L+2} \sim J_{4c} + A/(L+1)$  gives the extrapolated result  $J_{4c} = 0.05 \pm 0.01$  as the dashed line in Fig. 2. To test the precision of the value, we did the same analysis using the gap for  $\delta M = \pm 2$  instead of  $\Delta_{\pm}$  as solid squares shown in Fig. 2 where the fixed point can be obtained only for  $L \geq 10$ . It gave  $J_{4c} = 0.01 \pm 0.01$  which is not well coincide with the above result, which implies that the available system size is not enough to determine  $J_{4c}$  with the fitting of  $1/(L+1)$ . (Such a difficulty of the precise decision of the critical point by PRG is sometimes due to the logarithmic size correction in the case of the KT transition. [25]) Assuming that the system is gapless for  $J_4 = 0$ , we conclude  $J_{4c} = 0.05 \pm 0.04$  within the present analysis.

We present the GS magnetization curve in the thermodynamic limit for  $J_4 = 0$  and  $0.1$ . In the latter case the magnetization plateau should appear at  $m = \frac{1}{2}$  in contrast to the former, as discussed above. Note that the four-spin exchange interaction reduces the spin gap just above the nonmagnetic GS. According to our present analysis based on PRG, the gap for  $m = 0$  vanishes at a critical value  $\tilde{J}_{4c}$ , which should be distinguished from  $J_{4c}$  for  $m = \frac{1}{2}$ , and the nonmagnetic GS will belong to a different massive phase from the Haldane phase for  $J_4 > \tilde{J}_{4c}$ . The critical value  $\tilde{J}_{4c}$ , however, is obviously larger than  $0.1$ . Thus even in the case of  $J_4 = 0.1$  the spin gap due to the Haldane mechanism still exists for  $m = 0$  and we can use the same method to give the magnetization curve as used for the  $S = 1$  antiferromagnetic chain [28].

For  $J_4 = 0.1$  the left hand sides of the form (3) calculated for  $m = 0, \frac{1}{4}, \frac{1}{3}, \frac{1}{2}, \frac{2}{3}$  and  $\frac{3}{4}$  are plotted versus  $1/L$

in Fig. 3. It shows  $H_+ = H_-$  except for  $m = 0$  and  $\frac{1}{2}$ . Thus we take the mean value of the two for the magnetic field  $H$  for each  $m$ .

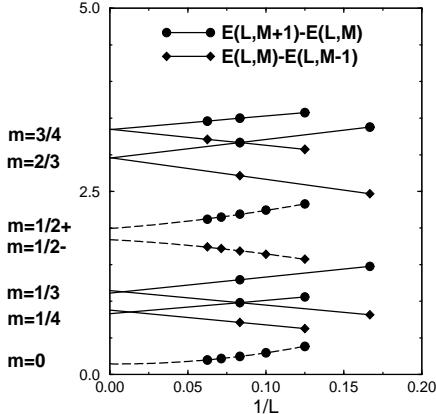


FIG. 3.  $E(L, M+1) - E(L, M)$  and  $E(L, M) - E(L, M-1)$  plotted versus  $1/L$  with fixed  $m$  for  $J_4 = 0.1$ . The extrapolated points for  $m = 0$ ,  $m = 1/2-$  and  $m = 1/2+$  correspond to the results of the Shanks' transformation  $H_{c1} = 0.15 \pm 0.03$ ,  $H_- = 1.84 \pm 0.06$  and  $H_+ = 1.99 \pm 0.09$ , respectively.

Since the nonmagnetic GS is massive for  $J_4 = 0$  and  $0.1$ , the size correction of  $H_+$  decays faster than  $\frac{1}{L}$  as shown in Fig. 3. Thus we use the Shanks' transformation [29]  $P'_n = (P_{n-1}P_{n+1} - P_n^2)/(P_{n-1} + P_{n+1} - 2P_n)$  twice for the sequence  $E(L, 1) - E(L, 0)$  for  $L = 6, 8, 10, 12$  and  $14$ , and obtain  $H_{c1} = 0.503 \pm 0.003$  and  $0.15 \pm 0.03$  for  $J_4 = 0$  and  $0.1$ , respectively. The saturation field  $H_{c2}$  is given by the  $L$ -independent quantity  $E(L, L) - E(L, L-1)$ .

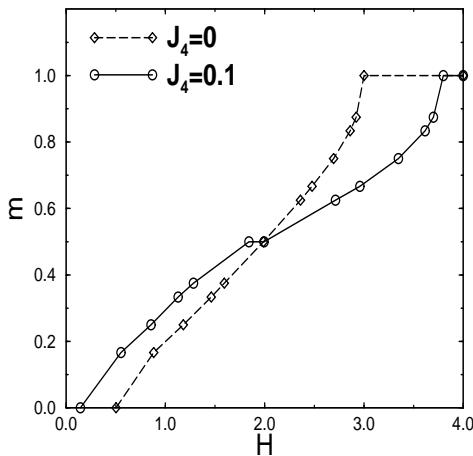


FIG. 4. GS magnetization curves in the thermodynamic limit for  $J_4 = 0$  and  $0.1$ . The latter has the magnetization plateau at half the saturation value.

In the case of  $J_4 = 0.1$ , for  $m = 1/2$   $H_+$  and  $H_-$  are

obviously different and the size correction decays faster than  $1/L$ , as shown in Fig. 3, which is consistent with a finite gap. Then we estimate  $H_+$  and  $H_-$  by the Shanks' transformation and get  $H_+ = 1.99 \pm 0.09$  and  $H_- = 1.84 \pm 0.06$ . For  $J_4 = 0$   $H_+$  and  $H_-$  correspond even at  $m = 1/2$ . We present the results for  $J_4 = 0$  and  $0.1$  in Fig. 4, where we also used the values of  $H$  for  $m = 1/6$ ,  $3/8$ ,  $5/8$ ,  $5/6$  and  $7/8$  which are estimated by the same method as mentioned above. The curve has a plateau at  $m = 1/2$  ( $H_- < H < H_+$ ) for  $J_4 = 0.1$ .

Our present PRG analysis shows that the gap does not behave as  $\Delta \sim (J_4 - J_{4c})^\nu$ . If we define the size-dependent exponent  $\nu_L$ , it diverges as  $L$  increases. Instead, if the gap behaves like

$$\Delta \sim \exp\left(-\frac{a}{(J_4 - J_{4c})^\sigma}\right), \quad (5)$$

as in the case of universality class of KT transitions ( $\sigma = \frac{1}{2}$ ), the Roomany-Wyld approximation for the Callen-Symanzik  $\beta$ -function [26], which is defined as

$$\beta_{L,L+2}(J_4) = \frac{1 + \log\left(\frac{\Delta_{L+2}(J_4)}{\Delta_L(J_4)}\right) / \log\left(\frac{L+2}{L}\right)}{\left[\frac{\Delta'_L(J_4)\Delta'_{L+2}(J_4)}{\Delta_L(J_4)\Delta_{L+2}(J_4)}\right]^{\frac{1}{2}}}, \quad (6)$$

should have the form

$$\beta_{L,L+2}(J_4) \sim (J_4 - J_{4c,L,L+2})^{1+\sigma}. \quad (7)$$

Fitting the form (7) to the calculated function (6) for each  $L$ ,  $\sigma$  is estimated as follows:  $\sigma_{10,12} = 0.38 \pm 0.10$ ,  $\sigma_{12,14} = 0.43 \pm 0.10$  and  $\sigma_{14,16} = 0.49 \pm 0.10$ . The results are consistent with  $\sigma = \frac{1}{2}$ . Thus we conclude the critical behavior near  $J_{4c}$  for  $m = \frac{1}{2}$  is characterized by the universality class of the KT transition.

Furthermore we estimate the central charge  $c$  of CFT, using the asymptotic form of the GS energy per site

$$\frac{1}{L} E(L, M) \sim \epsilon(m) - \frac{\pi}{6} c v_s \frac{1}{L^2} \quad (L \rightarrow \infty), \quad (8)$$

where  $v_s$  is the sound velocity which is the gradient of the dispersion curve at the origin. The result shown in Fig. 5 suggests  $c = 1$  with only a few percent errors for  $m = \frac{1}{2}$  and  $0 \leq J_4 \leq 0.1$ . It also supports the KT transition.

The critical exponent  $\eta$ , associated with the spin correlation function in the leg direction like  $\langle S_0^+ S_r^- \rangle \sim (-1)^r r^{-\eta}$ , can be estimated by the form of the gap  $\Delta_\pm \sim \pi v_s \eta / L$  ( $L \rightarrow \infty$ ) [2]. The estimated  $\eta$ , shown in Fig. 5, seems close to  $\frac{1}{2}$  around the critical point  $J_{4c}$ , rather than  $\frac{1}{4}$  which is expected for the KT transition. We think there is a possible jump from  $\eta = \frac{1}{4}$  to  $\eta = \frac{1}{2}$  at  $J_{4c}$  because the elementary excitations is expected to behave like the free Fermion systems ( $\eta = \frac{1}{2}$ ) at the edge of the plateau for  $J_4 > J_{4c}$  [5]. The present small cluster analysis could not detect such a discontinuity. Another exponent  $\eta^z$  defined as  $\langle S_0^z S_r^z \rangle \sim \cos(2k_F r) r^{-\eta^z}$  can also be estimated from the  $L$ -dependence of the soft mode

gap with the momentum  $2k_F = 2\pi m$  [21]. We checked the validity of the relation  $\eta\eta^z = 1$  around  $J_{4c}$  which is consistent with the Luttinger liquid theory [27] leading to  $\eta = \frac{1}{2}$  in the free Fermion case.

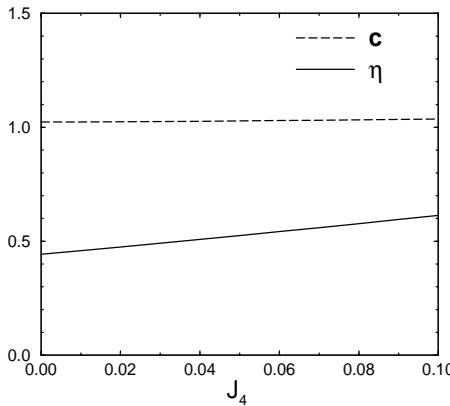


FIG. 5. Estimated central charge  $c$  and exponent  $\eta$  around  $J_{4c}$ . The result indicates  $c = 1$  which is consistent with the KT transition.  $\eta$  is close to  $\frac{1}{2}$  rather than  $\frac{1}{4}$ .

The spin gap at  $m = 0$  has already been observed in several real ladder compounds, for example  $\text{Cu}_2(\text{C}_2\text{H}_{12}\text{N}_2)_2\text{Cl}_4$  [30,20] and  $\text{La}_6\text{Ca}_8\text{Cu}_{24}\text{O}_{41}$  [31]. The magnetization plateau, however, has not been detected at any finite magnetization. We hope some new ladder materials with the field-induced spin gap will be discovered in the near future.

In summary the finite cluster calculation and size scaling study showed that the  $S = \frac{1}{2}$  antiferromagnetic spin ladder with the four-spin cyclic exchange interaction at every plaquette has the magnetization plateau at  $m = 1/2$  for  $J_4 > J_{4c} = 0.05 \pm 0.04$  and the phase transition with respect to  $J_4$  belongs to the KT universality class.

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